

NEA/NSC/DOC(2004)14
INDC(Ger)-050
Jül-4151

PROGRESS REPORT ON NUCLEAR DATA RESEARCH IN THE FEDERAL REPUBLIC OF GERMANY

for the Period April 1, 2003 to March 31, 2004



November 2004

Edited by
S.M. Qaim
Forschungszentrum Jülich GmbH
Institut für Nuklearchemie
Jülich, Federal Republic of Germany

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Berichte des Forschungszentrums Jülich; 4151

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FOREWORD

As in previous years, this report has been prepared to promote exchange of nuclear data research information between the Federal Republic of Germany and other member states of OECD/NEA and IAEA. It covers progress reports from the research centres at Karlsruhe and Jülich, and the universities of Dresden, Hannover and Köln. Each contribution is presented under the laboratory heading from where the work is reported. The names of other participating laboratories are also mentioned. The emphasis in the work reported here is on nuclear data for applied science programmes, such as those relevant to reactor technology and safety, transmutation concepts, accelerator shielding and development, astrophysical research, production of medically important radioisotopes, radiation therapy, etc.

The coordination of nuclear data activities at the international level is done by two committees: the NEA-Nuclear Science Committee (NEA-NSC) and the IAEA-International Nuclear Data Committee (INDC). The present Editor has the privilege and the responsibility of representing Germany in both the committees. This report therefore also serves as a background information to some areas of work of those committees.

Due to certain unforeseen circumstances the report is rather delayed this year. Nonetheless, it is hoped that it will still be of interest to the readers.

Jülich, November 2004

S.M. Qaim

This document contains information of a preliminary nature. Its contents should be used with discretion.

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**FORSCHUNGSZENTRUM KARLSRUHE
INSTITUT FÜR KERNPHYSIK**

1. An Optimized C₆D₆ Detector for Studies of Resonance-dominated (n,γ) Cross Sections*

R. Plag, M. Heil, F. Käppeler, P. Pavlopoulos¹, R. Reifarth², K. Wisshak and the _TOF collaboration

Hydrogen-free scintillators are indispensable for determining the small, resonance-dominated neutron capture cross sections of light and neutron magic nuclei, data which are needed for advanced reactor concepts, for transmutation of radioactive wastes as well as for astrophysical scenarios of neutron capture nucleosynthesis. A critical comparison of existing detector concepts by means of detailed GEANT simulations revealed unexpectedly large differences in neutron sensitivity. Based on these simulations an optimized detector was developed and successfully tested. Compared to a commercial detector the neutron sensitivity of this solution is more than an order of magnitude lower, thus allowing even extremely small capture/scattering ratios to be measured reliably.

* Nucl. Instr. Meth. A, 496 (2003) 425

¹ Astro-Particle Consortium, Nuclear Physics Laboratory, University of Ioannina, Greece and Department of Physics and Astronomy, Basel, Switzerland

² Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

2. New Experimental Validation of the Pulse Height Weighting Technique for Capture Cross Section Measurements*

U. Abbondanno et al.¹

The accuracy of the pulse height weighting technique for the determination of neutron capture cross sections is investigated. The technique is applied to measurements performed with C₆D₆ liquid scintillation detectors of two different types using capture samples of various dimensions. The data for well known (n,γ) resonances are analysed using weighting functions obtained from Monte Carlo simulations of the experimental set-up. Several causes of systematic deviation are identified and their effect is quantified. In all these cases the reaction yield agrees with the standard value within 2%.

* Nucl. Instr. Meth. A, 521 (2004) 454

¹ The _TOF collaboration, see <http://pceet075.cern.ch/>

3. Safety of Food Irradiation with High Energy X-rays: Experimental Evidence *

O. Grégoire¹, M.R. Cleland¹, J. Mittendorfer², S. Dababneh³, D.A.E. Ehlermann⁴,
F. Käppeler³, M. Knörr⁴, J. Logar⁵, J. Meissner⁶, B. Mullier¹, F. Stichelbaut¹, D.W. Thayer⁷

The radiological safety of red meat irradiated with 7.5 MeV X-rays has been investigated theoretically and verified by dedicated experiments. The experimental setup has been chosen to simulate closely the situation in a commercial irradiation facility. Samples of meat and meat ash were located in a large volume of fresh meat at the position of the highest neutron fluence and irradiated to 15 kGy, twice the maximum dose allowed by the US FDA for meat irradiation. The induced activity was measured with high purity germanium (HPGe) gamma ray spectrometers. In order to evaluate the safety of treatment with any kind of electron accelerators, 2 experiments have been performed with different accelerators delivering electrons with a narrow and a broad energy spread.

The measured activities and theoretical estimates are in the same order of magnitude. An evaluation of the corresponding radiation exposure has been compared to natural background and the extent of possible biological effects has been considered. The paper concludes that food irradiation with X-rays from 7.5 MeV electrons, even with a broad energy spectrum, can be regarded safe from the standpoint of public health.

* *Radiat. Phys. Chem.* 67 (2003) 169

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⁷ Agricultural Research Service, US Department of Agriculture, East Mermaid Lane 600, Wyndmoor, PA 19038, USA

4. Irradiation of Medical Devices with High Energy X-rays: Experimental Tests with Electrons in a Broad Energy Spectrum.

O. Grégoire¹, M.R. Cleland¹, J. Mittendorfer², S. Dababneh³, D.A.E. Ehlermann⁴,
F. Käppeler³, M. Knörr⁴, B. Mullier¹, F. Stichelbaut¹

The induced radioactivity in medical devices when sterilized with high energy X-rays has been theoretically and experimentally addressed in a previous publication. However, those tests and calculations had been performed for monoenergetic electrons striking the X-ray target. In order to verify the safety of X-ray irradiation with 7.5 MeV electrons, we have to consider different acceleration devices, even those producing a broad energy spectrum. The present experimental tests have been performed with such an accelerator.

Metallic samples, identified in the previous experiment as those producing the most important activation, have been irradiated in a large volume of cellulose. The activity was measured with a highly sensitive assembly of spectrometers and compared to activation results with X-rays produced by monoenergetic electrons.

Measured activities are well below reporting limits in Europe and the United States, and the corresponding radiation exposure of workers is below the maximum dose for exemption of

authorization. It is therefore likely that the sterilization process, whatever the equipment used, can be exempted as a practice.

* Radiat. Phys. Chem. 68 (2003) 149

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⁴ Bundesforschungsanstalt für Ernährung, Haid-und-Neu-Strasse 9, 76131 Karlsruhe, Germany

5. Gamma Spectroscopy using two Clover Detectors in Close Geometry

S. Dababneh¹, N. Patronis², P.A. Assimakopoulos², J. Görres³, M. Heil¹, F. Käppeler¹, D. Karamanis², S. O'Brien³, R. Reifarth⁴

A gamma spectrometer composed of two Clover Ge detectors in close geometry is described. The advantages and drawbacks of the different modes of operation are investigated. The use of offline coincidence analysis for substantial background reduction is presented and an experimental approach for the determination of the summing correction factor is formulated.

* Nucl. Instr. Meth. A, 517 (2004) 230

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³ University of Notre Dame, Department of Physics, Notre Dame, IN 46556, USA

⁴ Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

6. Measurement of the n TOF Beam Profile with a Micromegas Detector *

J. Pancin et al.¹

A Micromegas detector was used in the neutron Time-Of-Flight (n_TOF) facility at CERN to evaluate the spatial distribution of the neutron beam as a function of its kinetic energy. This was achieved over a large range of neutron energies by using two complementary processes: at low energy by capture of a neutron via the ⁶Li(n,α)t reaction, and at high energy by elastic scattering of neutrons on gas nuclei (argon+isobutane or helium+isobutane). Data are compared to Monte Carlo simulations and an analytical function fitting the beam profile has been calculated with a sufficient precision to use in neutron capture experiments at the n_TOF facility.

* Nucl. Instr. Meth. A, 524 (2004) 102

¹ the _TOF collaboration, see <http://pceet075.cern.ch/>

7. A Low Background Neutron Flux Monitor for the n_TOF Facility at CERN *

S. Marrone^{1,2}, P.F. Mastinu³, U. Abbondanno⁴, R. Baccomi⁴, E. Boscolo Marchi³, N. Bustreo³, N. Colonna¹, F. Gramegn^{1, 3}, M. Loriggiol^{1, 3}, S. Marigo³, P.M. Milazzo⁴, C. Moreau⁴, M. Sacchetti¹, G. Tagliente¹, R. Terlizzi^{1,2}, G. Vannini⁵, G. Aerts⁶, E. Berthomieux⁶, D. Cano-Ott⁷, P. Cennini⁸, C. Domingo-Pardo⁹, L. Ferrant¹⁰, E. Gonzalez-

Romero⁷, F. Gunsing⁶, M. Heil¹¹, F. Kaeppler¹¹, T. Papaevangelou¹², C. Paradela¹³, P. Pavlopoulos¹⁴, L. Perrot¹⁰, R. Plag¹¹, J.L. Tain⁹, H. Wendler⁸

A small-mass system has been developed for monitoring the flux of neutrons with energy up to 1 MeV at the new time-of-flight facility at CERN, n TOF. The monitor is based on a thin Mylar foil with a ^6Li deposit, placed in the neutron beam, and an array of silicon detectors, placed outside the beam, for detecting the products of the $^6\text{Li}(n,\alpha)^3\text{H}$ reaction. The small amount of material on the beam ensures a minimal perturbation of the flux and minimizes the background related to scattered neutrons. Moreover, a further reduction of the γ -ray background has been obtained by constructing the scattering chamber hosting the device in carbon fibre. A detailed description of the flux monitor is presented, together with the characteristics of the device, in terms of efficiency, resolution and induced background. The use of the monitor in the measurement of neutron capture cross-sections at n_TOF is discussed.

* Nucl. Instr. Meth. A, 517 (2004) 389

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¹³ Universidade de Santiago de Compostela, Spain

¹⁴ Pôle Universitaire Léonard de Vinci, Paris La Défense, Paris, France

8. Stellar He Burning of ^{18}O : A Measurement of Low-energy Resonances and their Astrophysical Implications *

S. Dababneh¹, J. Görres², M. Heil¹, F. Käppeler¹, H. Leiste¹, R. Reifarth³, M. Wiescher²

The $^{22}\text{Ne}(\alpha,n)$ reaction is the main neutron source for neutron capture nucleosynthesis (s process) in massive stars and plays also a significant role for the s process in thermally pulsing asymptotic giant branch stars. In these scenarios, ^{22}Ne is produced by the reaction sequence $^{14}\text{N}(\alpha,\gamma)^{18}\text{F}(\beta^+)^{18}\text{O}(\alpha,\gamma)^{22}\text{Ne}$. While the first reaction is well understood, α capture on ^{18}O was affected by considerable uncertainties. At the temperatures of stellar He burning, the reaction rate is determined by two resonances at α energies of 470 and 566 keV. Since these resonances were not yet successfully measured, the rates had to be based on estimated resonance strengths. In the present work, the first direct measurement of the partial strengths of these extremely weak low-energy resonances is reported. The use of a high-efficiency segmented Ge detector in coincidence with bismuth germanate oxide counters covering a large solid angle led to a significantly improved experimental sensitivity, thus allowing for the clear identification of specific γ transitions. As a result, resonance strengths of $0.71 \pm 0.17 \mu\text{eV}$ and $0.48 \pm 0.16 \mu\text{eV}$ could be obtained for the 566- and 470-keV resonances, respectively. When compared to the previously reported upper limits of 1.7 μeV , these results provide a reliable basis for the determination of the reaction rate during stellar He burning. Accordingly, these data reduce the uncertainties in the s process neutron balance.

* Phys. Rev. C 68 (2003) 025801

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9. ⁹⁵Mo(n,α) Cross Section from 1 eV to 500 keV: A Test of the α+nucleus Optical Potential used in Calculating Reaction Rates for Explosive Nucleosynthesis *
W. Rapp^{1,2}, P. E. Koehler,¹ F. Käppeler,² and S. Raman^{1,†}

We have measured the ⁹⁵Mo(n,α) cross section in the energy range from 1 eV to 500 keV at the Oak Ridge Electron Linear Accelerator ORELA. This work is part of a series of (n,α) measurements for deriving a reliable global α+nucleus potential, which is an essential ingredient in nuclear statistical model calculations of the reaction rates for unstable nuclei involved in explosive *p*-process nucleosynthesis. The ⁹⁵Mo(n,α) rate shows a strong sensitivity to the α+nucleus potential used in the calculations, and therefore these new data should be very useful in obtaining an improved potential. For example, although there is a factor of 5 variation in the rate calculated using different potentials, an older model and a newer one using one of three recently proposed potentials are in good agreement with our new data.

* Phys. Rev. C 68 (2003) 015802

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[†] deceased

10. Neutron Capture Cross Section of ¹³⁹La *

S. O'Brien¹, S. Dababneh², M. Heil³, F. Käppeler³, R. Plag³, R. Reifarth⁴, R. Gallino⁵, M. Pignatari⁵

The neutron capture cross section of ¹³⁹La has been measured relative to that of ¹⁹⁷Au by means of the activation method. The sample was irradiated in a quasi-stellar neutron spectrum for *kT* = 25 keV generated via the ⁷Li(*p,n*)⁷Be reaction with the proton energy adjusted 30 keV above the threshold. Maxwellian averaged neutron capture cross sections were calculated for energies *kT* = 5 to 100 keV. The new value for *kT* = 30 keV is found to be 31.6 ± 0.8 mb, 18% lower and considerably less uncertain than the previously recommended value of 38.4 ± 2.7 mb. With these results the *s*- and *r*-process components could be more accurately determined, making lanthanum a reliable *s*- and *r*-process indicator in stellar spectroscopy.

* Phys. Rev. C 68 (2003) 035801

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- 11. Evidence for $M1$ Scissors Resonances Built on the Levels in the Quasicontinuum of ^{163}Dy**
M. Krtička¹, F. Bečvár¹, J. Honzátko², I. Tomandl², M. Heil³, F. Käppeler³, R. Reifarth⁴, F. Voss³, K. Wisshak³

Spectra of two-step γ cascades following the thermal $^{162}\text{Dy}(n, \gamma)^{163}\text{Dy}$ reaction have been measured. Distinct peak-like structures observed at the midpoints of these spectra are interpreted as a manifestation of the low-energy isovector $M1$ vibrational mode of excited ^{163}Dy nuclei.

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FORSCHUNGSZENTRUM KARLSRUHE INSTITUT FÜR REAKTORSICHERHEIT

1. Measurement and Analysis of the Activation of Eurofer Steel in an IFMIF-like Neutron Spectrum

P. Bém*, V. Burjan*, U. Fischer, M. Götz*, M. Honusek*, V. Kroha*, J. Novák*, S. P. Simakov, E. Šimečková*

An activation experiment on the Eurofer low activation steel was performed at the cyclotron of the Nuclear Physics Institute (NPI), Řež [1], with the objective to provide the experimental data base for validating activation cross-section data in the energy range relevant for the International Fusion Material Irradiation Facility (IFMIF). An IFMIF-like white neutron field extending up to 32 MeV neutron energy was produced by bombarding a flowing heavy water target with a 37 MeV proton beam. The irradiation of Eurofer-97 steel samples was performed at two different locations with distances to the source of 1.7 mm and 47 mm and mean neutron flux densities up to 10^{11} and $10^{10}/\text{cm}^2/\text{s}$, respectively. The induced gamma radioactivity was determined by gamma ray spectrometry at different cooling time intervals using calibrated HPGe detectors. Specific activities in units of Bq per kg of the sample were determined for 21 different product radionuclides.

Computational pre- and post-analyses of the Eurofer activation experiment have been performed at the Forschungszentrum Karlsruhe with the objective to check and validate activation cross-section data in the energy range relevant to IFMIF. A detailed 3D geometry model of the experimental set-up including the heavy water target irradiated by a proton beam, the activation foils and their holders was developed for Monte Carlo simulations with the MCNPX code [2]. It was shown that both the LA-150h proton cross-sections and the MCNPX built-in reaction models are not capable of reproducing the p-D₂O neutron source term with sufficient accuracy. As a consequence, the neutron flux spectrum at the positions of the Eurofer foils was constructed by combining the measured spectrum above 5 MeV neutron energy at large distance from the target with the one calculated by MCNPX below this energy. The absolute normalisation has been performed relative to the yield of ²⁴Na generated in the Al monitoring foils.

The activation analysis of the Eurofer steel has been performed with the ALARA inventory code [3] and activation cross-sections from the Intermediate Energy Activation File IEAF-2001 [4] which is applicable for neutron energies up to 150 MeV. The gamma ray yields of 21 radionuclides generated in the Eurofer steel were finally calculated and compared with the measurements. Results of the activation analysis are shown in Fig. 1 in terms of C/E (calculation/experiment) ratios of the resulting γ -activities. For many of the radionuclides generated in low energy threshold nuclear reactions such as ⁴⁸Sc, ⁵⁴Mn, ⁵⁹Fe, ⁵¹Cr, ¹⁸²Ta, the agreement is quite

* Nuclear Physics Institute, Řež, Czech Republic

satisfactory. Large discrepancies have been found for the radio-nuclide inventories produced in nuclear reactions with thresholds greater than 10 MeV (e. g. ^{48}V , ^{48}Cr , ^{51}Cr , ^{52}Mn , ^{56}Co , ^{57}Ni , ^{177}Ta). These results indicate the need for updating the relevant IAEA-2001 cross sections in the high energy domain.

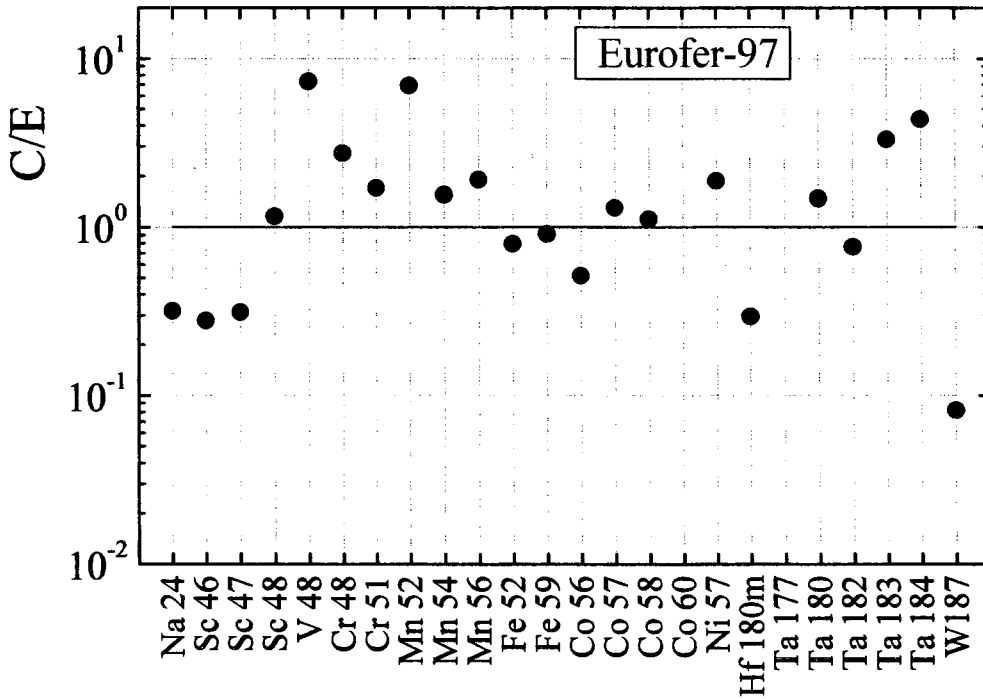


Fig. 1 Comparison of calculated (ALARA/IEAF-2001) and measured [1] radioactivities induced by irradiating Eurofer steel in an IFMIF-like white neutron field.

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2. Evaluation of $n + {}^{184}\text{W}$ Cross Section Data up to 150 MeV

P. Pereslavytsev, U. Fischer

In the European Fusion Technology Programme, highest priority has been assigned to high quality data evaluations for the tungsten isotopes. This includes the neutron energy range above 20 MeV to enable intermediate energy calculations of the IFMIF (International Fusion Irradiation Materials Irradiation Facility) neutron source facility. As a first step for the preparation of an EFF/JEFF high energy general purpose data library, an evaluation for $n + {}^{184}\text{W}$ reaction system up to 150 MeV was performed [1]. The GNASH code was used for the calculation of nuclear reaction cross sections and the ECIS code for the neutron elastic and direct inelastic scattering cross sections. Recent high energy experimental data were taken into account for evaluating the total and non-elastic cross sections. A set of optical model potentials up to 150 MeV was chosen on the basis of detailed comparisons of available experimental data and calculations with the ECIS and the SCAT2 code. Both global and local potentials for neutrons, protons, deuterons, tritons and alphas were considered. To improve the neutron emission spectra, collective excitations were also included in GNASH. Double-differential cross sections of the emitted particles were calculated on the basis of the Kalbach systematics.

Results of ECIS95 coupled channel calculations for the differential elastic scattering cross sections of ${}^{\text{nat}}\text{W}$ and ${}^{184}\text{W}$ are shown in Fig. 2. It is noted that both the Young deformed potential and the Koning global spherical potential can reproduce the experimental data rather well.

A complete data file in ENDF6 data format was prepared for ${}^{184}\text{W}$ covering the full energy range data up to 150 MeV. As a general rule, the ENDF/B-VI.8 data were adopted below 20 MeV with some modifications to achieve agreement with recent experimental data. The data file was successfully processed with NJOY/ACER and checked through MCNP calculations for a tungsten benchmark experiment.

The evaluation of high energy W cross-section data will be concluded for the other stable W isotopes using the same approach as for the evaluation of $n + {}^{184}\text{W}$ cross sections.

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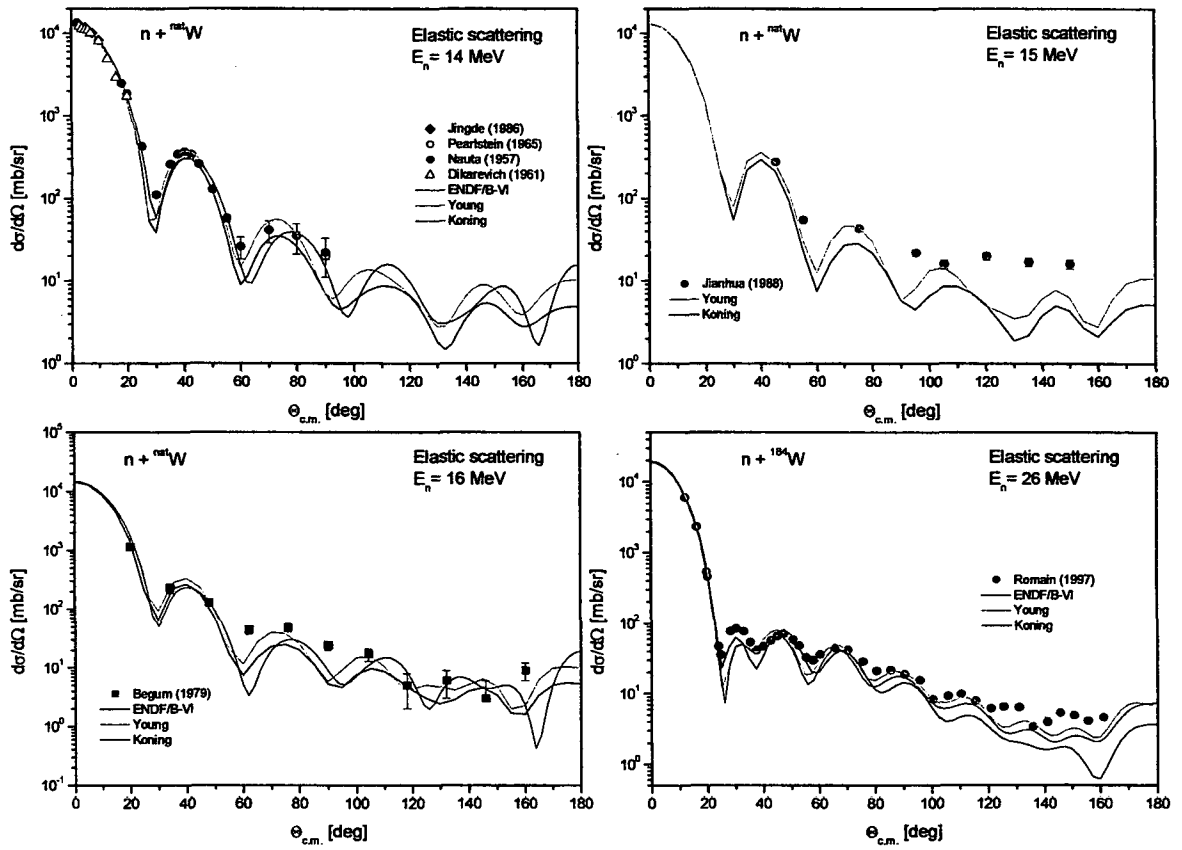


Fig. 2 Differential elastic scattering cross section of ${}^{nat}\text{W}$ and ${}^{184}\text{W}$ as a function of scattering angle: Comparison of experimental data and calculations using different optical model potentials (Young, Koning & Delaroche, ENDF/B-VI)

INSTITUT FÜR NUKLEARCHEMIE FORSCHUNGSZENTRUM JÜLICH

1. Fundamental Studies on Isomeric Cross Sections

S. M. Qaim, S. Sudár[†], Ye. Skakun[§], A. Fessler^{**}

In order to study the effect of reaction channel on the isomeric cross-section ratio, the formation of the isomeric pair $^{52\text{m,g}}\text{Mn}$ was investigated in four nuclear reactions, namely, $^{52}\text{Cr}(\text{p},\text{n})$ -, $^{52}\text{Cr}({}^3\text{He},\text{t})$ -, $^{54}\text{Fe}(\text{d},\alpha)$ - and $^{54}\text{Fe}({}^3\text{He},\alpha\text{p})$ -processes. The metastable state has a spin 2^+ and the ground state 6^+ . Both of them decay by $\text{EC} + \beta^+$ emission, i.e. there is almost no isomeric transition to the ground state. For each reaction the standard stacked-foil technique of excitation function measurement was used. Thin samples were prepared by electrolytic deposition. Irradiations were done at the compact cyclotron CV28 of the FZ Jülich and the beam currents were measured via monitor reactions. The radioactivity of each product was determined using HPGe detector γ -ray spectrometry. Due to the relatively low cross section for the formation of $^{52\text{g}}\text{Mn}$ at low incident particle energies, the product was radiochemically separated. The results for the $^{52}\text{Cr}(\text{p},\text{n})^{52\text{m,g}}\text{Mn}$ and $^{54}\text{Fe}(\text{d},\alpha)^{52\text{m,g}}\text{Mn}$ reactions have been reported earlier [cf. Klein, Rösch, Qaim, *Radiochimica Acta* **88**, 253 (2000); Zaman, Spellerberg, Qaim, *Radiochimica Acta* **91**, 105 (2003)]; the other results are new.

From the individual cross sections the isomeric cross-section ratios were deduced. All the ratios were plotted against the incident projectile energy and the results are shown in Fig. 1. For each reaction, initially the ratio decreases, but becomes almost constant at high excitation energies. It is interesting to note that the (p,n) and $({}^3\text{He},\text{t})$ reactions (curves A and B) occur on the same target nucleus and the product studied is also the same. The same is true for the $({}^3\text{He},\alpha\text{p})$ and (d,α) reactions (curves C and D). The magnitudes of the isomeric cross-section ratios differ considerably. The observed trends show clearly that the reaction channel influences the isomeric cross-section ratio considerably.

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Further similar studies have been performed on the isomeric pair $^{102m,g}\text{Rh}$, the metastable state having a spin (2^-) and the ground state (6^+). The reactions studied were $^{103}\text{Rh}(p,p'n)^{102m,g}\text{Rh}$, $^{101}\text{Ru}(^3\text{He,pn})^{102m,g}\text{Rh}$ and $^{102}\text{Ru}(^3\text{He,t})^{102m,g}\text{Rh}$. The data analysis has been completed and nuclear model calculations are being carried out.

With a view to studying the formation of relatively high-spin isomers in nuclear reactions, measurements have been done on the pairs $^{195m,g}\text{Hg}$ and $^{197m,g}\text{Hg}$ in $^{nat}\text{Pt}(^3\text{He,xn})$ - and $^{nat}\text{Pt}(\alpha,\text{xn})$ -reactions from their thresholds up to 36 MeV in the case of ^3He -particles and 28 MeV for α -particles. In each case the metastable state has the spin ($13/2^+$) and the ground state ($1/2^-$). Further analysis of the data and nuclear model calculations are under way.

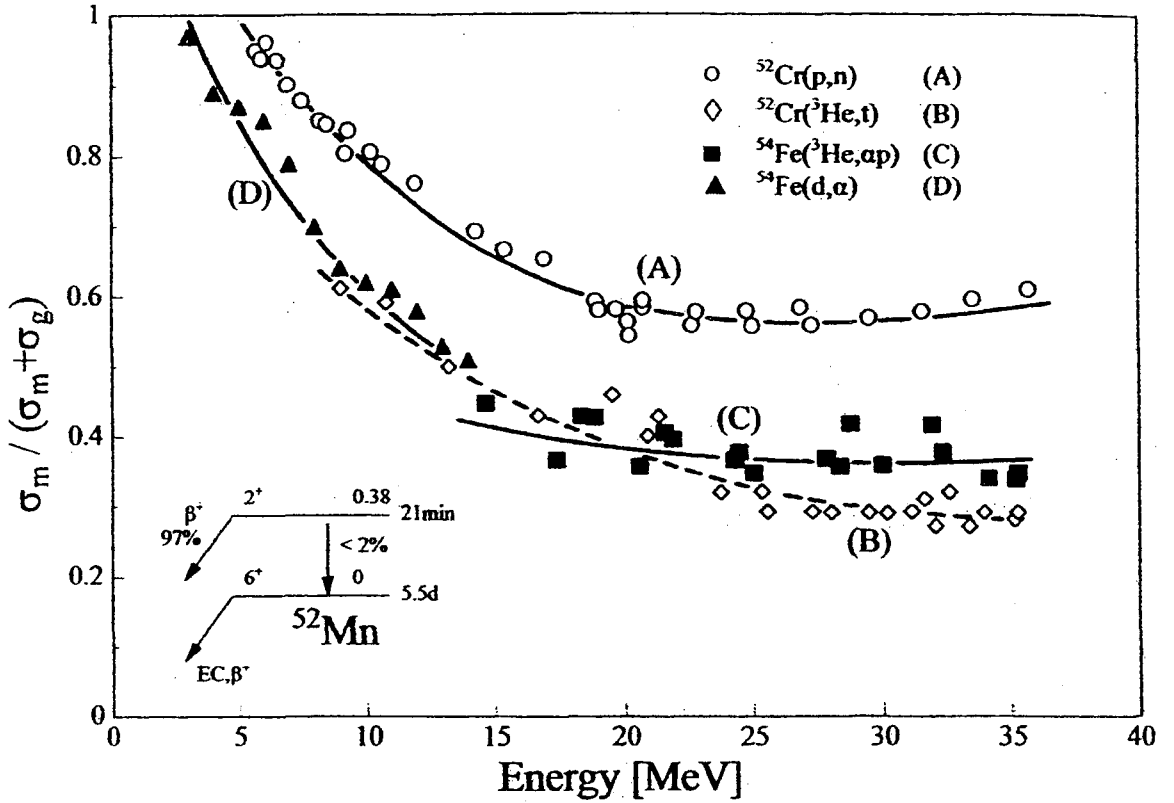


Fig. 1 Experimental isomeric cross-section ratios for the formation of $^{52m,g}\text{Mn}$ in several charged particle induced reactions, plotted as a function of incident particle energy.

2. Nuclear Reaction Cross Section Data for Medical Applications

As in previous years, systematic studies on charged particle induced reaction cross sections for medical applications were continued. During the period of the present Progress Report following investigations were carried out.

a) *Cross sections and yields of nuclear processes for the production of the positron emitters ^{76}Br , ^{124}I and ^{82}Sr (^{82}Rb)*

B. Scholten, H.E. Hassan*, S. Spellerberg, D. Steyn**, N. van der Walt**, S. Takács⁺⁺, F.Tárkányi⁺⁺, S.M. Qaim, H.H. Coenen

The positron emitting radionuclide ^{76}Br ($T_{1/2} = 16.0$ h) is of considerable medical interest for use in Positron Emission Tomography (PET). For its production the reaction $^{76}\text{Se}(p,n)^{76}\text{Br}$ is commonly used; its database, however, was relatively weak in the energy range below 15 MeV. Cross sections were measured for this reaction via the stacked-foil technique using highly enriched ^{76}Se as target material and high resolution HPGe detector γ -ray spectrometry. The results are shown in Fig. 2. The new data define the reaction threshold precisely; they should allow an accurate calculation of the expected ^{76}Br yield at low-energy cyclotrons. The calculated ^{76}Br yield over the energy range $E_p = 15 \rightarrow 8$ MeV, for example, amounts to 360 MBq/ $\mu\text{A}\cdot\text{h}$.

For ^{76}Br production a completely new route, namely $^{78}\text{Kr}(d,\alpha)^{76}\text{Br}$, was also investigated. Highly enriched ^{78}Kr gas was irradiated in a thin-walled cylinder and the activities of the products were determined via γ -ray spectrometry. The results are shown in Fig. 3. The ^{76}Br obtained is of high radionuclidic purity. However, due to the relatively low cross section of the (d, α) process, the yield of ^{76}Br is relatively low. Over the energy range of $E_d = 14 \rightarrow 8$ MeV, for example, the ^{76}Br yield amounts to only 60 KBq/ $\mu\text{A}\cdot\text{h}$. The method is therefore not suitable for routine production of ^{76}Br .

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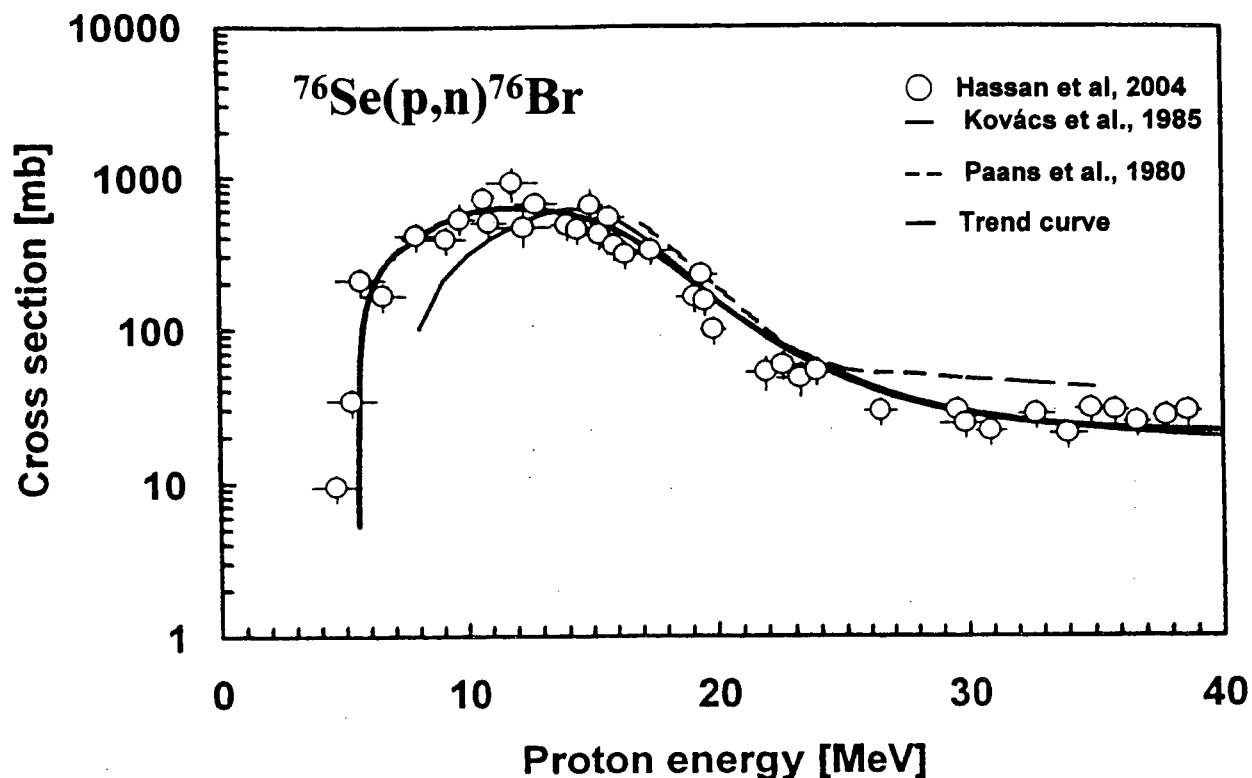


Fig. 2 Excitation function of the $^{76}\text{Se}(p,n)^{76}\text{Br}$ reaction, determined using highly enriched ^{76}Se as target material.

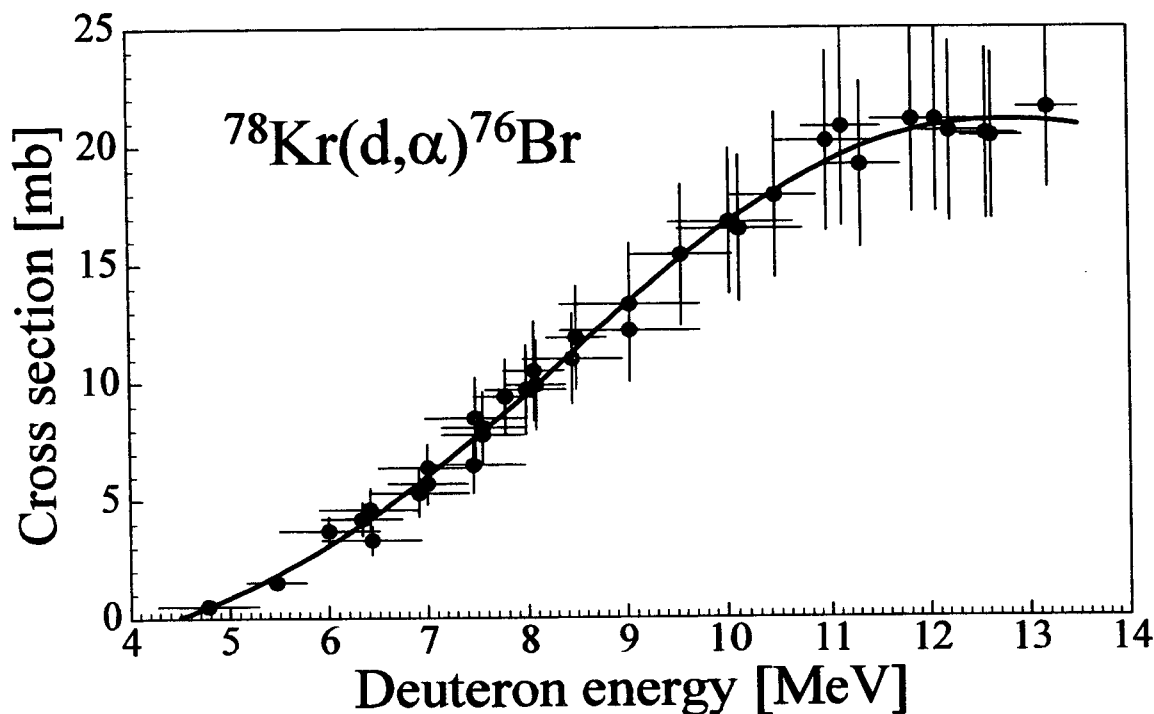


Fig. 3 Excitation function of the $^{78}\text{Kr}(d,\alpha)^{76}\text{Br}$ reaction, determined using highly enriched ^{78}Kr gas as target material.

The positron emitting radionuclide ^{124}I ($T_{1/2} = 4.18$ d) is both a diagnostic and a therapeutic nuclide and finds application in PET studies. Because of its increasing significance, a continuous search for its alternative methods of production is under way. Extensive studies on the $^{126}\text{Te}(p,3n)^{124}\text{I}$ reaction started last year were completed over the energy range of 8 to 70 MeV. The yield of ^{124}I via this route appears to be much higher but the levels of the impurities need to be critically analysed.

The radionuclide ^{82}Sr ($T_{1/2} = 25.3$ d) is the parent of the positron emitting short-lived ^{82}Rb ($T_{1/2} = 1.3$ min) which is widely used in PET studies. The parent, needed for preparing a generator system which finds application at PET centres without a cyclotron, can be produced via several routes. In recent years the $^{85}\text{Rb}(p,4n)^{82}\text{Sr}$ reaction has been commonly used; an evaluation by the IAEA, however, showed some discrepancies in the available data. Recently, two new detailed studies on this reaction were completed [cf. Kastleiner, Qaim, Nortier, Blessing, van der Walt, Coenen, *Appl. Radiat. Isot.* **56**, 685 (2002); Ido, Hermanne, Ditroi, Szücs, Mahunka, Tárkányi, *Nucl. Instr. Methods B* **194**, 369 (2002)]. All the data were now analysed and evaluated, and a reliable excitation function curve was obtained. In order to validate those data, thick RbCl targets, covering various energy ranges between 44 and 66 MeV, were irradiated under well-defined conditions. In each case, the experimental thick target yield of ^{82}Sr was determined and compared with the value deduced from the evaluated curve. The agreement was within 10 %. These thick target yield measurements thus validated the evaluated curve which can now be used as a standard in ^{82}Sr production studies.

b) Cross sections for production of the therapeutic radionuclides ^{103}Pd , ^{140}Nd , ^{169}Yb and ^{192}Ir

K. Hilgers, I. Spahn, Ye. Skakun^{*}, F. Tárkányi⁺, S. Takács⁺, S.M. Qaim, H. H. Coenen

The radionuclide ^{103}Pd ($T_{1/2} = 17.0$ d) is of considerable significance in brachytherapy. The studies on the $^{102}\text{Ru}(^3\text{He},2n)^{103}\text{Pd}$ and $^{100}\text{Ru}(\alpha,n)^{103}\text{Pd}$ reactions, using highly enriched isotopes as target materials, mentioned in the last Progress Report have now been completed. From the experimental excitation function of each reaction the yield of ^{103}Pd

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was calculated. The results for the above mentioned two reactions are given in Fig. 4 together with the yields obtained via other nuclear processes. Evidently the $^{103}\text{Rh}(\text{d},2\text{n})^{103}\text{Pd}$ reaction gives the highest yield but, due to the non-availability of very high deuteron currents, the $^{103}\text{Rh}(\text{p},\text{n})^{103}\text{Pd}$ reaction is most commonly used. The present results show that under certain circumstances the $(^3\text{He},2\text{n})$ and (α,n) reactions can serve as alternative routes for the production of ^{103}Pd .

The radionuclide ^{140}Nd ($T_{1/2} = 3.4$ d) is a potentially useful therapeutic isotope since it emits several Auger electrons per decay. For its production two routes, namely $^{\text{nat}}\text{Ce}(^3\text{He},\text{xn})^{140}\text{Nd}$ and $^{141}\text{Pr}(\text{p},2\text{n})^{140}\text{Nd}$, have been suggested. During the period of this Progress Report detailed cross-section measurements on both the reactions were completed. A comparison of the two routes is given in Table 1. The $(\text{p},2\text{n})$ reaction has a higher yield. In both cases a medium-sized cyclotron is needed.

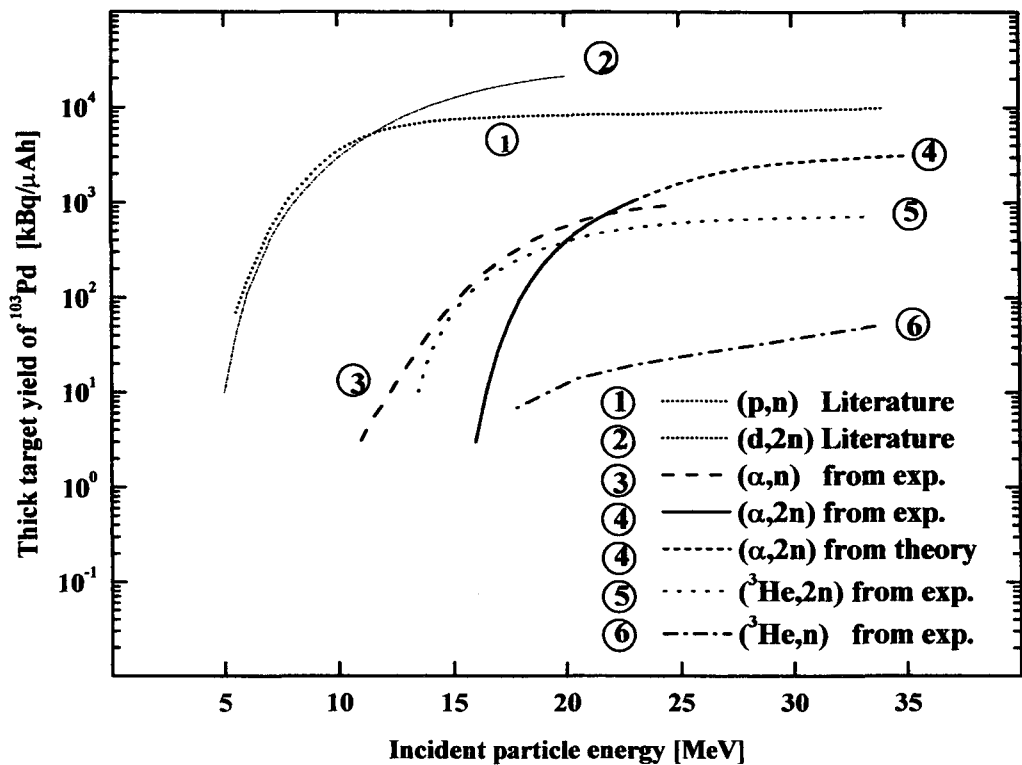


Fig. 4 Integral yields of ^{103}Pd calculated from the excitation functions of $(^3\text{He},2\text{n})$ and (α,n) reactions studied in this work and (p,n) and $(\text{d},2\text{n})$ reactions reported in the literature. The $(^3\text{He},\text{n})$ and $(\alpha,2\text{n})$ processes were also investigated in this work; their yields at energies available at the CV 28 are, however, low.

Table 1 Comparison of production routes of ^{140}Nd

Production route	E_{max} [MeV]	σ_{max} [mb]	Suitable energy range [MeV]	Integral yield [MBq/ $\mu\text{A}\cdot\text{h}$]
$^{\text{nat}}\text{Ce}(^3\text{He},\text{xn})^{140}\text{Nd}$	27	800	$35 \rightarrow 20$	22
$^{141}\text{Pr}(\text{p},2\text{n})^{140}\text{Nd}$	23	1300	$28 \rightarrow 15$	250

The radionuclides ^{169}Yb ($T_{1/2} = 32.0$ d) and ^{192}Ir ($T_{1/2} = 78.8$ d) are important therapeutic isotopes, the former an Auger electron emitter and the latter a low-energy β^- emitter. As far as applications are concerned, the former is an emerging radionuclide but the latter is more established. Both of them are produced in nuclear reactors via the (n,γ) reaction. With a view to increasing the specific activity of the product, and under a Research Agreement with the IAEA, we studied the excitation functions of the $^{169}\text{Tm}(\text{p},\text{n})^{169}\text{Yb}$ and $^{192}\text{Os}(\text{p},\text{n})^{192}\text{Ir}$ reactions from their respective thresholds up to 40 MeV in the case of the former reaction and up to 20 MeV in the latter. The data analysis for the first reaction is in progress. The results for the $^{192}\text{Os}(\text{p},\text{n})^{192}\text{Ir}$ reaction are shown in Fig. 5. Measurements were done using thin samples of enriched ^{192}Os , prepared via electrolytic deposition on Ni, and high-resolution γ -ray spectrometry. The shape of the excitation function (Fig. 5) is similar to that of other (p,n) -reactions in this mass region. However,

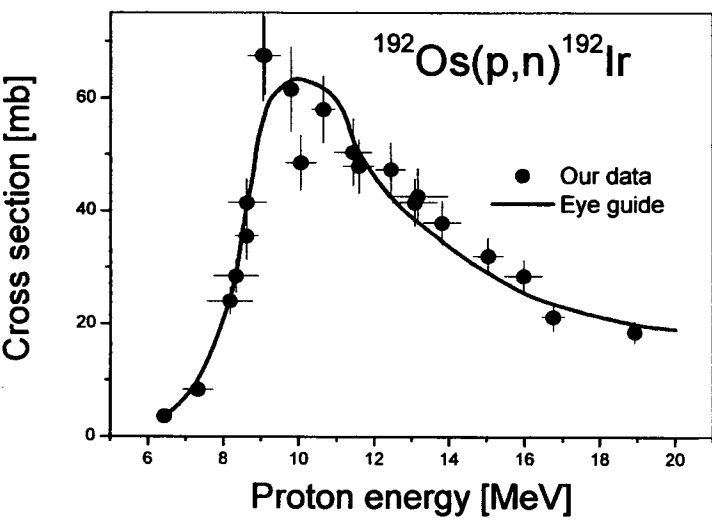


Fig. 5 Excitation function of the $^{192}\text{Os}(\text{p},\text{n})^{192}\text{Ir}$ reaction, determined using enriched ^{192}Os as target material.

the magnitude is rather low. From the given curve an ^{192}Ir yield of $0.16 \text{ MBq}/\mu\text{A}\cdot\text{h}$ is obtained in the energy range $E_p = 16 \rightarrow 8 \text{ MeV}$. Due to the relatively low yield, it is predicted that the batch yield of this radionuclide in cyclotron production would not reach the same level as in reactor production. The specific activity of the product, however, would be much higher.

c) *Proton therapy related activation cross sections*

K. Kettern, B. Scholten, S. Spellerberg, D. Steyn*, N. van der Walt*, Yu. N. Shubin⁺, S.M. Qaim, H. H. Coenen

In proton therapy, the activation cross section data are of limited interest. Two aspects, however, need some consideration.

- (i) Formation of short-lived β^+ emitters in human tissue
- (ii) Formation of long-lived activation products in collimator materials.

Regarding the short-lived β^+ emitters, the on-going measurements on the formation of ^{11}C ($T_{1/2} = 20.3 \text{ min}$) and ^{13}N ($T_{1/2} = 10.0 \text{ min}$) in the interactions of protons of up to 200 MeV with the elements carbon, nitrogen and oxygen were completed. The results were compared with those from the hybrid model calculations using the code ALICE-IPPE. An accurate description was found to be difficult because of the non-statistical properties of light nuclei caused by their structure effects. Thus standard recommended input parameters could not be used. However, after some adjustments of the parameters based on experimental data, a satisfactory agreement between theory and experiment was obtained.

Activity calculations using the measured excitation functions showed that the contribution of the positron emitters ^{11}C and ^{13}N to the activation of human tissue generated by therapeutic proton beams is not negligible. Detailed results will be presented later.

As far as activation of collimator materials is concerned, we concentrated on two elements, viz. titanium and lead. Extensive data on the formation of short-lived activation products in the interactions of protons of energies up to 200 MeV with the two materials

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were available. However, some soft radiation emitting longer-lived products like ^{45}Ca ($T_{1/2} = 163$ d, $E_{\beta^-} = 300$ keV), ^{49}V ($T_{1/2} = 330$ d, K_{α} X-rays = 4.5 keV) and ^{204}Tl ($T_{1/2} = 3.78$ a, $E_{\beta^-} = 760$ keV) had not been investigated. In the present work clean radiochemical separations were performed and the radioactivity was determined via low-level β^- counting, liquid scintillation counting or X-ray spectrometry. The results for the $^{\text{nat}}\text{Ti}(p,x)^{45}\text{Ca}$ process are given in Fig. 6. It is interesting that the nuclear model calculation using the code ALICE-IPPE reproduces the experimental excitation function well.

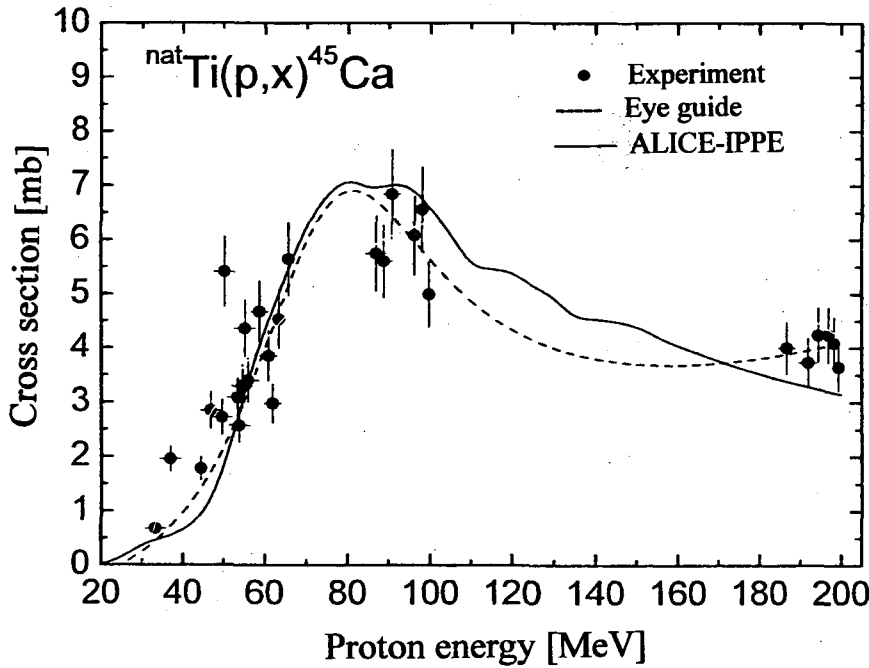


Fig. 6 Excitation function of the $^{\text{nat}}\text{Ti}(p,x)^{45}\text{Ca}$ process.

d) Compilation and preliminary evaluation of data

B. Scholten, S. Sudár*, F. Tárkányi⁺, S.M. Qaim

The Institute signed a Research Agreement with the IAEA in 2002 to participate in the CRP on Nuclear Data for Therapeutic Radionuclides, undertaking the responsibility to evaluate nuclear data for four nuclear processes, viz. $^{64}\text{Ni}(p,n)^{64}\text{Cu}$, $^{70}\text{Zn}(p,\alpha)^{67}\text{Cu}$, $^{103}\text{Rh}(p,n)^{103}\text{Pd}$ and $^{124}\text{Te}(p,n)^{124}\text{I}$. The compilation of data was completed, the missing data were introduced in the EXFOR file and a preliminary screening of the data was done. The final evaluation will be done by a theory oriented evaluator.

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3. Other Activities

a) Chair of INDC

Syed M. Qaim continued to act as Chairman of the International Nuclear Data Committee (INDC) of the IAEA. The term of office will cover the period up to the end of 2006.

b) NRC-6

The Institute organised the 6th International Conference on Nuclear and Radiochemistry (NRC-6) in Aachen, Germany, from 29 August to 03 September 2004. More than 300 scientists from over 35 countries participated. Nuclear data activities constituted an integral part of the programme of the conference.

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INSTITUT FÜR KERN- UND TEILCHENPHYSIK TECHNISCHE UNIVERSITÄT DRESDEN

1. Measurement and Analysis of Radioactivity induced in CuCrZr by D-T Neutrons^{*}

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CuCrZr alloys (0.754 wt-% Cr, 0.163 wt-% Zr) are candidate materials for the heat sink in the first wall of the blanket and in the divertor of fusion devices. The radioactivity in a fusion reactor is mainly produced by fast neutrons of the 14 MeV fusion peak, where the number of open reaction channels is much higher than by thermal neutrons, and for some reactions the cross section is large in this energy region. In the present work the γ -ray activities induced by the 14 MeV neutrons in this near-plasma material were investigated.

For planning the irradiation experiment, a calculation was made with European Activation System (EASY, [1]). The material was assumed to be irradiated under reactor conditions, i. e. with a flux density of the 14 MeV neutrons corresponding to a power density of 1.0 MW/m², for a period of one year. The results obtained for the contact dose rate as a function of the decay time after irradiation are shown in Fig.1 for a range up to 10⁴ years. The safety-related short-term radioactivity is expected to be determined by the isotopes ⁶²Cu and ⁶⁴Cu. The recycling limit and the hands-on-limit of the material depend on the ⁶⁰Co activity. The long-term radioactivity which is important for waste management mainly depends on the ²⁶Al activity. It is produced by the ²⁷Al(n,2n)²⁶Al reaction. Its dose rate is well below the hands-on-limit, if the Al impurity is 0.0059 wt-% as found in the analyses [2].

Fig. 1 shows that measurements of the γ -activities in the range of decay time labelled by t_m (shown by shading) permit testing the calculated radioactivity induced by 14 MeV neutrons up to the recycling limit.

^{*} Work supported by the European Fusion Development Agreement (Task TTMN-002)

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³ Coordination Centre "Atomsafety", Sergiev Posad, Russia

⁴ Russian Research Centre "Kurchatov Institute", Moscow, Russia

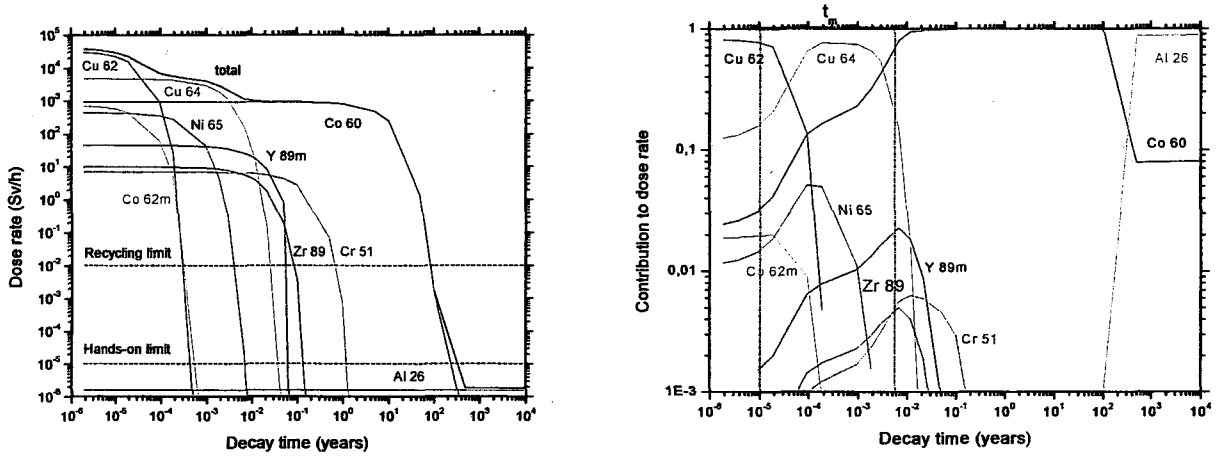


Fig. 1 Calculated contact dose rate (left hand) and contribution of the different radionuclides to the total dose rate (right hand) after irradiation of CuCrZr with fusion peak neutrons of 1.0 MW/m^2 power density for one year as a function of the decay time.

The irradiation of the CuCrZr sample (mass 0.8334 g) was performed at a D-T neutron generator of TUD. During 203.0 min a total neutron fluence of $2.1 \cdot 10^{12} \text{ cm}^{-2}$ was applied. It was measured by simultaneous activation of niobium foils and evaluating the activity induced by the $^{93}\text{Nb}(n,2n)^{92\text{m}}\text{Nb}$ reaction. A possible background component of thermal neutrons was checked by $^{197}\text{Au}(n,\gamma)$ activation using thin foils. The ratio of thermal to 14 MeV neutrons was found to be less than 10^{-5} . The attenuation of the neutron and γ -ray fluxes in the sample as well as the geometry relations source-sample and sample-detector were determined by 3D Monte Carlo calculation.

γ -ray spectra were taken with an HPGe-spectrometer at a distance of 50 mm between the irradiated sample and the detector at several decay times in the range labelled with t_m in Fig.1. The γ -activities identified by energy and half-life were used to determine the nuclide activities using γ -yield data from JEF2.2 [3].

The measured activities were analysed with the versions EASY-2001 and EASY-2003. The results of the analysis for individual activities are presented in Table 1. The uncertainties of the calculated activities ($\Delta C/C$) include both cross section and half-life errors, as estimated by EASY. The uncertainties of the experimental values ($\Delta E/E$) take into account the possible errors of the γ -activity measurements (statistical uncertainty of the γ -counting, the uncertainty

of the efficiency calibration of the spectrometer including the geometry factor), sample mass, γ -yield data and the neutron flux monitoring.

Table 1 Results obtained for the activity of radionuclides;

radionuclides identified, their half-life and the γ -rays used to determine the activity, the neutron reactions producing these radionuclides, the ratios of calculated-to-experimental activity (C/E), obtained with EASY-2001 and EASY-2003, and the uncertainties of both the calculated ($\Delta C/C$; EASY 2001) and the experimental activities ($\Delta E/E$).

Radio-nuclide	Half-life	E_γ (keV)	Reaction Contribution (%)	C/E EASY- 2001	C/E EASY- 2003	$\Delta C/C$ (%)	$\Delta E/E$ (%)
^{51}Cr	27.7 d	320.2	$^{52}\text{Cr}(n,2n)$ 100	1.16	1.16	5.	10.8
^{60}Co	5.27 y	1332.5 1173.2	$^{63}\text{Cu}(n,\alpha)$ 100	1.14	1.12	48.6	9.7
^{61}Co	1.65 h	320.2	$^{65}\text{Cu}(n,n'\alpha)$ 100	1.73	1.09	20.	9.1
$^{62\text{m}}\text{Co}$	13.9 min	1163 2004	$^{65}\text{Cu}(n,\alpha)$ 100	1.53	1.53	62.	11.4
^{65}Ni	2.5 h	1481.8 1116.1 366.5	$^{65}\text{Cu}(n,p)$ 100	1.19	1.19	10.	7.4
^{62}Cu	9.74 min	1163 875.7	$^{63}\text{Cu}(n,2n)$ 100	1.09	1.09	5.	11.3
^{64}Cu	12.71 h	1345.9	$^{65}\text{Cu}(n,2n)$ 99.4 $^{63}\text{Cu}(n,\gamma)$ 0.6	1.15	1.15	5.	34.8
^{89}Zr	3.27 d	909.2	$^{90}\text{Zr}(n,2n)$ 100	1.12	1.12	51.	10.3

The radionuclides of Table 1 are produced each by one dominant reaction. Ratios of calculated-to-experimental activity (C/E) that deviate from unity by more than 50% and that are outside the experimental error bars were obtained with EASY-2001 for ^{61}Co and $^{62\text{m}}\text{Co}$. The validation process of the EASY cross section data [4] results in better agreement with the measured activity of ^{61}Co for EASY-2003, whereas the discrepancy for $^{62\text{m}}\text{Co}$ remains unchanged. For the other activities, C and E agree within the total uncertainties. Smaller $\Delta E/E$ values than the $\Delta C/C$ in some cases indicate possibilities for further improvements on the EASY data base. More details can be found in Ref. [5].

Summarizing, it can be stated that the recycling limit of the material after irradiation under fusion power plant conditions (power density of 1 MW/m², exposition of 1 year) will be reached after about 100 years. All activities that are important up to the recycling limit were experimentally determined and compared with the calculated values. From the ratio of calculated-to-experimental total activity it may be concluded that the expected activation performance is validated in this time range within 15%.

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2. Validation Experiment on γ -Activities of Y irradiated in Fusion Peak Neutron Field

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Oxide dispersion strengthened steels (ODS) are primary candidates as materials with a potential for use at high temperatures under strong neutron flux environments, like fusion DEMO plant first wall and blanket structural material. The characteristic feature of ODS steels is to introduce Y₂O₃ particles into the matrix of ferretic/martensic steels, which serve as a block for mobile dislocations to improve the high-temperature strength, and as a sink of point defects induced by radiation displacement to maintain superior radiation resistance.

For structural materials the radioactivity induced by neutrons is a central safety related topic. The short-term radioactivity with half-lives in the range of minutes to weeks is of interest for the heat production and the shut-down dose rate, whereas the long-term radioactivity in the range up to several hundred years is interesting for waste management. Most of the steels used in fusion design, like AISI316-IG, F82H and EUROFER, have already been investigated. In the present work pure yttrium was irradiated, to investigate the activation performance of this new constituent in the ODS steels.

In a calculation with the European Activation System [1], this material was assumed to be irradiated under power plant conditions; this means with a flux density of 14 MeV neutrons

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corresponding to a power density of 1.0 MW/m^2 , for a period of one year. The results obtained for the contact dose rate as a function of the decay time after irradiation are shown in Fig.1.

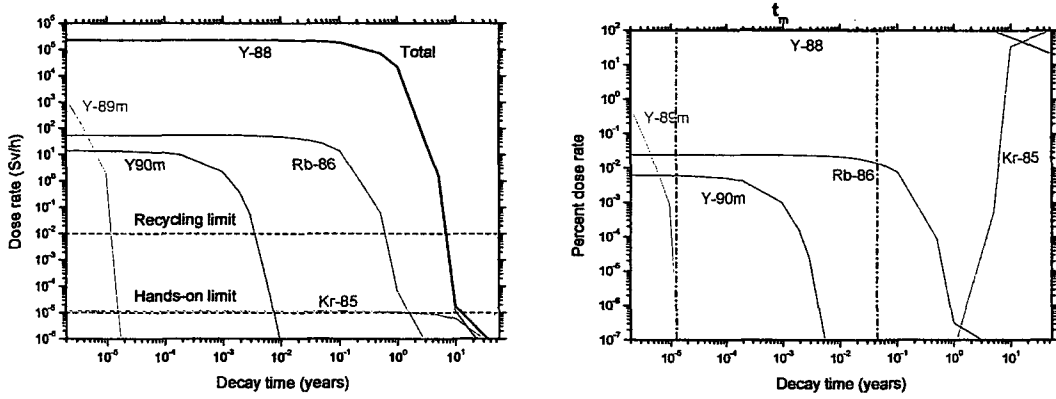


Fig. 1 Calculated contact dose rate (left hand) and contribution of the different radionuclides to the total dose rate (right hand) after irradiation of Y with fusion peak neutrons of 1 MW/m^2 power density for one year as a function of the decay time.

The dominant radionuclide is ^{88}Y up to the hands-on limit which is reached in about 10 years. $^{89\text{m}}\text{Y}$, $^{90\text{m}}\text{Y}$ and ^{86}Rb contribute only to a very small extent. To measure the radioactivity induced by neutrons in Y, γ -ray spectra were taken from an irradiated sample at decay times in the region labelled as t_m in Fig. 1. The sample had a thickness of 0.64 mm, a circular front area with a diameter of 25 mm and a mass of 1.3850 g. The irradiation of the sample was performed at a neutron generator of TUD. During $t_{\text{irr}} = 229 \text{ min}$ a total neutron fluence of $1.179 \cdot 10^{11} \text{ cm}^{-2}$ was applied. It was determined by simultaneous activation of niobium foils and evaluation of the activity induced by the $^{93}\text{Nb}(n,2n)^{92\text{m}}\text{Nb}$ reaction.

γ -ray spectra were taken with an HPGe-spectrometer at a distance of 50 mm to the irradiated sample. The γ -activities identified by energy and half-life were used to determine the nuclide activities using γ -yield data from EASY. The attenuation of the neutron and γ -ray fluxes in the sample as well as the geometry factors (source-sample and sample-detector) were determined by 3D Monte Carlo calculations.

The measured activities were analysed with the version EASY-2003 [1] of the European Activation System. The results of the analysis are presented in Table 1. The uncertainty of the calculated activities ($\Delta C/C$) includes both cross section and half-life errors as estimated by EASY-2003. The uncertainty of the experimental values ($\Delta E/E$) takes into account the possible errors of the γ -activity measurements (statistical uncertainty of the gamma-ray

counting, the uncertainty of the efficiency determination of the spectrometer including the geometry factor) sample mass, γ -yield data and the neutron flux monitoring. More details on the experiment and cross section analyses can be found in Ref. [2].

Table 1 Results of irradiation and analysis;
 radionuclides identified, their half-lives and γ -rays with yield data used to determine the activity, the neutron reaction producing the nuclide, the ratio of calculated-to-experimental activity (C/E) and the uncertainty of both the calculated and the experimental activity.

Nuclide	Half-life	E_{γ} (keV)	Y_{γ} (%)	Reaction Contribution (%)	C/E	$\Delta C/C$ (%)	$\Delta E/E$ (%)
^{88}Y	106.6 d	898.02 1836.01	94 99.36	$^{89}\text{Y}(n, 2n)$ 100	1.13	5.0	9.4
^{90m}Y	3.19 h	202.51 479.53	95.81 89.98	$^{89}\text{Y}(n, \gamma)$ 100	1.14	50.0	11.7

The measured activities nearly agree with the calculated values within the uncertainty range. In the case of ^{90m}Y the deviation from unity is approximately covered by the experimental uncertainty. This result may be used to improve the relatively large uncertainty estimation of the EASY library data.

Summarizing, it can be stated that the hands-on limit of Y after irradiation under fusion power plant conditions (power density of 1 MW/m², exposition of 1 year) will be reached after about 10 years. During this time the only dominant radionuclide is ^{88}Y . The activity measured for ^{88}Y in the present experiment agrees with the calculated value within 13%, and the activation performance calculated with EASY-2003 is validated at this level. For the ^{90m}Y activity experimental and calculated values agree within 14%.

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Production of Residual Nuclides at Medium-Energies – Finalizing the HINDAS Project

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Introduction

The project HINDAS “High- and Intermediate-Energy Data for Accelerator-Driven Systems” [1] was initiated in the European 5th Framework with the goals to establish a comprehensive and consistent data base covering all aspects of nuclear reaction data relevant at intermediate and high energies for accelerator-driven technologies and to develop two new code systems for the modeling of nuclear reactions. Including sub-contractors, nearly 20 European institutions collaborated within the HINDAS project. Three target elements, Fe, Pb, and U, were chosen for which a complete experimental database was and is being established by which the newly developed models should be scrutinized. The HINDAS project made use of accelerators at UCL Louvain-La-Neuve, TSL University of Uppsala, PSI Villigen, KVI Groningen, GSI Darmstadt, FZJ Jülich, and the PSI/ETH Tandem AMS Facility at ETHZ Zürich to cover energies from 20 MeV up to 2 GeV.

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In 2003, the HINDAS project was completed. We here give an overview about some results of the HINDAS work-package 3 which dealt with measurements of cross sections for residual nuclide production by p- and n-induced reactions for energies between 30 MeV and 200 MeV, of cross sections for n-induced fission for energies between 30 and 180 MeV, of excitation functions for the production of long-lived and stable rare gas isotopes by p-induced reactions from thresholds up to 2.6 GeV, and the investigations of fission cross sections.

In addition to the work for the HINDAS project, measurements of excitation functions for the production of cosmogenic nuclides in extraterrestrial matter and respective model calculations were continued [2-7].

Residual nuclide production by proton-induced reactions

Under the HINDAS project the cross section data base developed by our group in recent years was extended to the heavy target elements Ta, W, Pb, and Bi [8 – 12]. As a by-product of these investigations the half-life of ^{205}Bi was revised [13]. New cross sections were also measured for the production of 19 radionuclides from the target element iron from thresholds up to 2.6 GeV. For the target element lead a comprehensive set of excitation functions published recently [8] was completed by AMS-measurements of cross sections for the production of the long-lived radionuclides ^{10}Be , ^{36}Cl , and ^{129}I [14 – 16] and by mass spectrometric measurements of stable and radioactive rare gas isotopes of He, Ne, Ar, Kr, and Xe [17, 18]. Irradiation experiments with natural uranium were performed at the injector cyclotron at PSI and about 400 cross sections for the production of 18 radionuclides for energies from 21 MeV to 69 MeV were measured [19]. Not too many cross sections were available in the literature for the proton-induced production of residual nuclides from uranium. Moreover, they were mostly old and neither systematic nor comprehensive with respect to the product nuclide and energy coverage. To improve this situation, further collaborations were initiated during the HINDAS project. First results on the production of residual nuclides from natural uranium by proton-induced reactions at 600 MeV are available [20] and evaluations of previous irradiation experiments on thorium and uranium at Saturne for energies between 200 MeV and 2.6 GeV are underway.

All these data allow for stringent tests of nuclear models and codes when calculating cross sections for residual nuclide production from thresholds up to 2.6 GeV. The new codes are TALYS for energies up to about 200 MeV and INCL4+ABLA for higher energies (upcoming HINDAS final re-

port). Comprehensive tests of the new codes, TALYS and INCL4+ABLA for Fe, W, Ta, Pb, Bi, and U for energies from thresholds up to 2.6 GeV were performed [12]. Fig. 1 gives, as an example, the excitation function for the production of $^{52m+g}\text{Mn}$ from iron, and fig. 2 for the production of ^{205}Bi from bismuth.

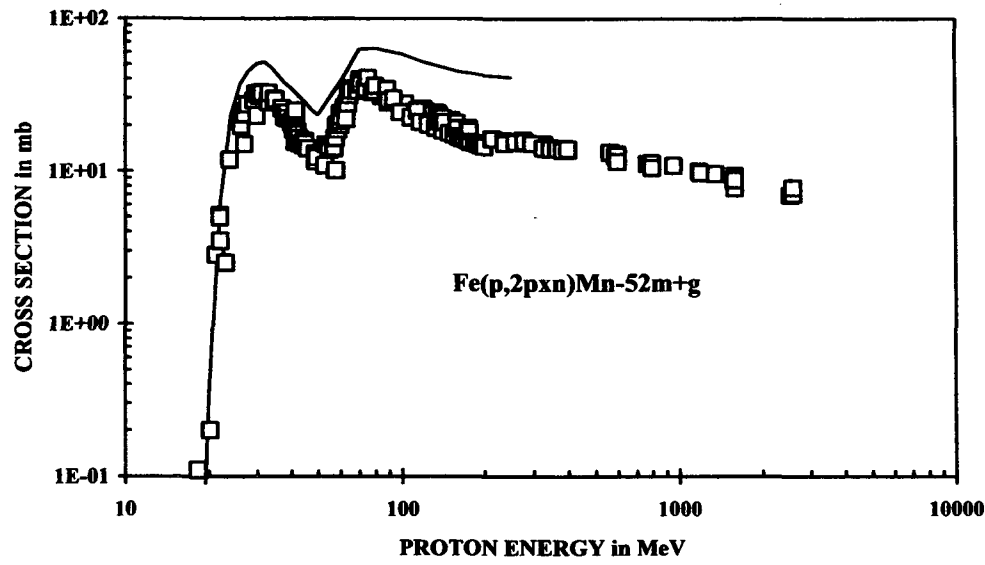


Fig. 1: Comparison of experimental cross sections from this work and previous work of our group for the reaction $^{nat}\text{Fe}(p,2pxn)^{52m+g}\text{Mn}$ with theoretical ones calculated by the TALYS code.

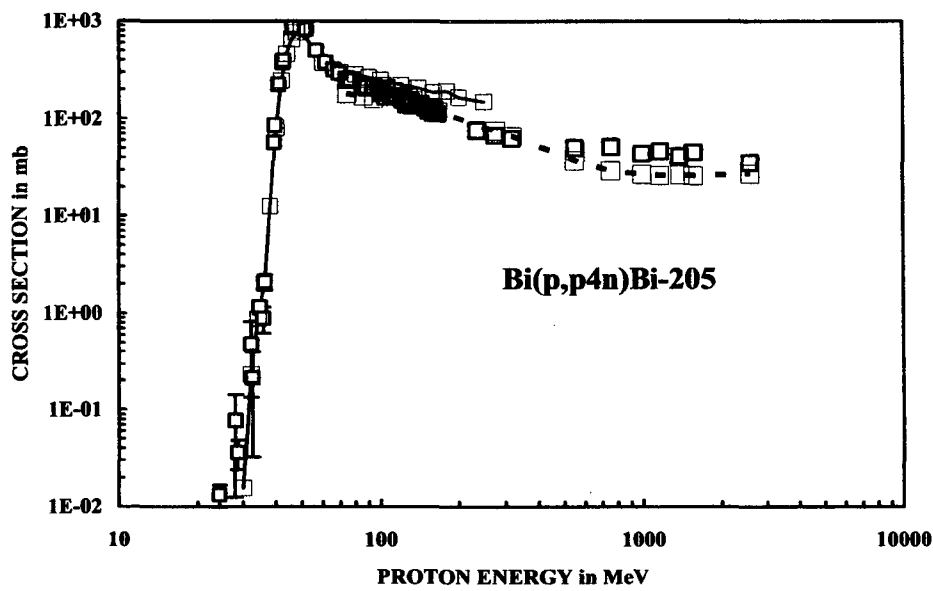


Fig. 2: Production of ^{205}Bi from bismuth by proton-induced reactions. Comparison of experimental results (squares) with theoretical cross sections calculated with INCL4+ABLA (broken line) and TALYS (solid line).

Long-lived residual nuclide production has to be considered as essential recording to waste management problems in ADS systems. The cross sections for the production of most of these nuclides are scarcely known. During the HINDAS project the excitation functions for the proton-induced production of the long-live radionuclides ^{129}I ($T_{1/2} = 1.6 \times 10^7 \text{ a}$), ^{36}Cl ($T_{1/2} = 3.0 \times 10^5 \text{ a}$), ^{26}Al ($T_{1/2} = 7.2 \times 10^5 \text{ a}$) and ^{10}Be ($T_{1/2} = 1.6 \times 10^6 \text{ a}$) from natural lead were measured using AMS at ETH H  nggerberg [14 – 16]. Fig. 3 presents the cross sections for the production of ^{10}Be from lead. ^{10}Be is formed as a fragmentation product, a reaction mode for which no reaction model exists up to now.

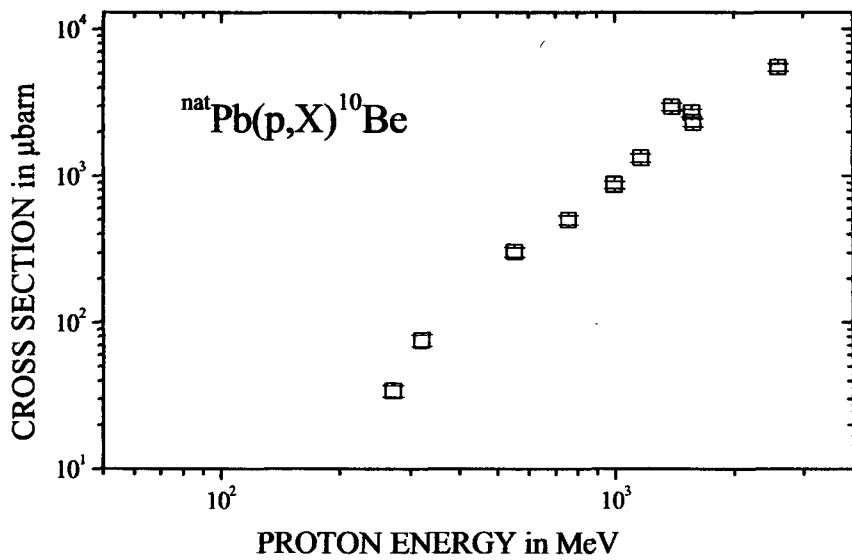


Fig. 3: Excitation function for production of ^{10}Be in proton-irradiation of $^{\text{nat}}\text{Pb}$.

The production of noble gas isotopes in lead by proton-induced reactions is of special importance for design studies of accelerator driven systems and energy amplifier. Such measurements were performed within the HINDAS project by direct measurement of light gas isotopes under the NESSI collaboration. Furthermore, in order to test the consistency with earlier mass spectrometric measurements the production of stable and radioactive rare gas isotopes of He-, Ne-, Ar-, Kr-, and Xe from natural lead by proton-induced reactions was investigated from threshold up to 2.6 GeV by rare gas mass spectrometry. Apart from some exceptions, the database for the proton-induced production of noble gas isotopes from lead is consistent and nearly complete. In contrast to the production of He from Al and Fe, where the cross sections obtained by thin-target irradiation experiments are up to a factor of 2 higher than the NESSI data, both datasets agree for the He production from lead (Fig. 4).

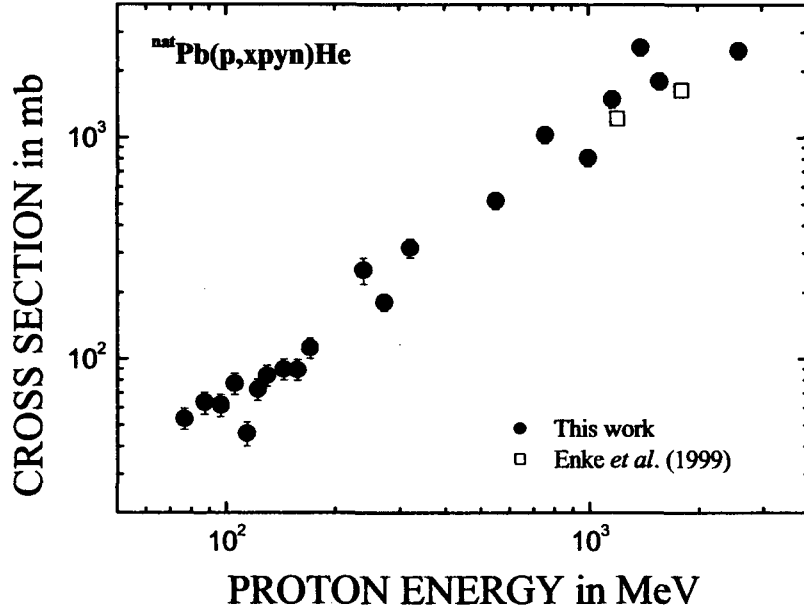


Fig. 4: Excitation function for the production of total He, *i.e.* the direct production of ^3He and ^4He , from lead by proton-induced reactions. Open squares are data from the NESSI collaboration [21].

Residual nuclide production by neutron-induced reactions

Within the HINDAS project, activation experiments with quasi mono-energetic neutrons produced by the $^7\text{Li}(p,n)^7\text{Be}$ reaction were performed at the neutron beam lines at TSL [22, 23] and UCL [24] in order to determine excitation functions for the production of residual radionuclides from a variety of target elements up to 175 MeV.

A total of 10 activation experiments covered proton energies between 36.4 and 178.8 MeV at UCL and TSL. The target elements C, N, O, Mg, Al, Si, Fe, Co, Ni, Cu, Ag, Te, Pb, and U were irradiated. Residual radionuclides with half-lives between 20 min and 5 years were measured by off-line γ -spectrometry. Information on the energy dependence of the neutron spectra in the targets was obtained by modeling the neutron spectra by Monte Carlo techniques using the LAHET/MCNP code system [25, 26]. These transport calculations started either from the experimentally determined neutron spectra (at UCL) or from the systematics of experimentally measured neutron emission spectra of the $^7\text{Li}(p,n)$ -reaction [27] (at TSL). The calculations described the transport of the neutrons into

the target stacks and into the individual targets as well the production and transport of secondary particles inside the massive target stacks which cannot be neglected [4].

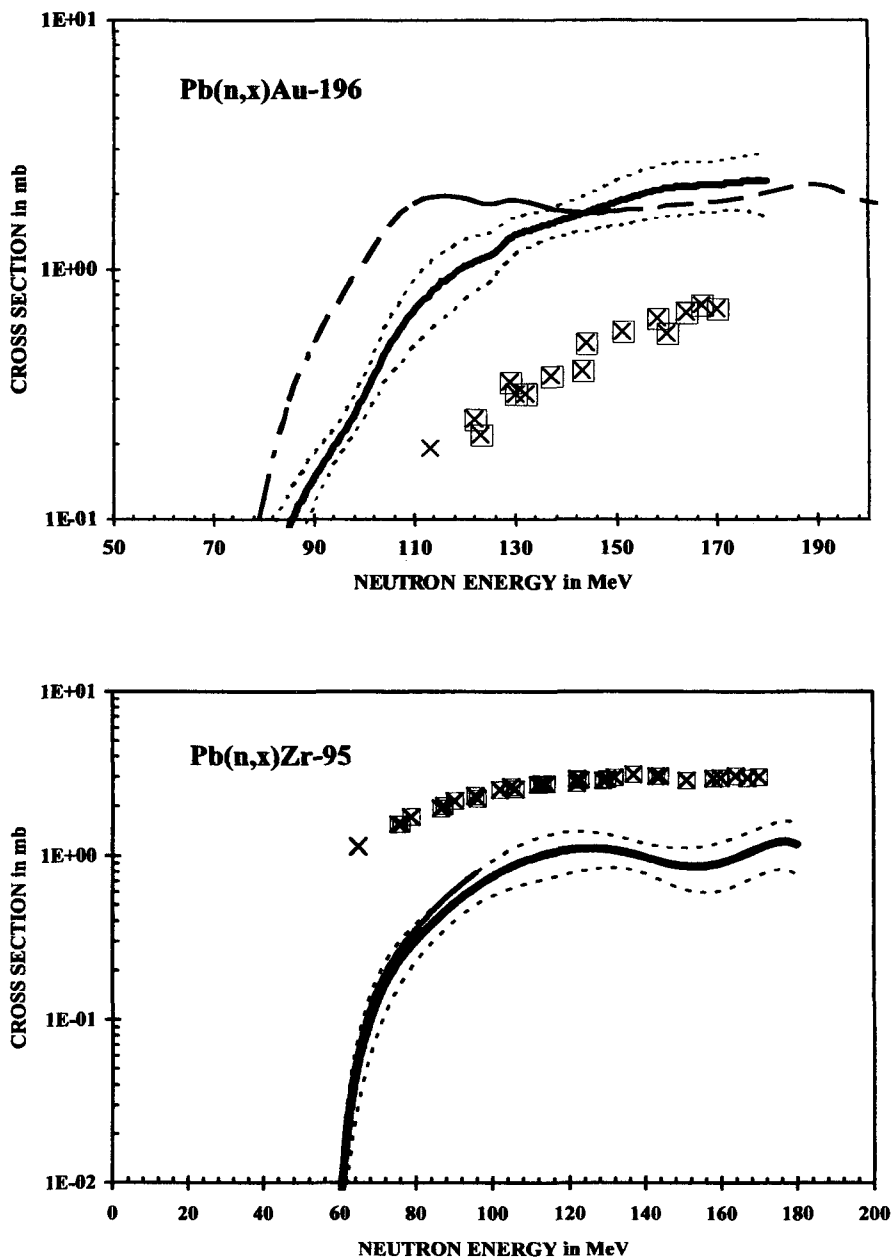


Fig. 5: Unfolded cross sections (solid lines) with their uncertainties (shaded areas) for $^{nat}\text{Pb}(n,x)^{196}\text{Au}$ and $^{nat}\text{Pb}(n,f)^{95}\text{Zr}$ compared with the respective cross sections from [8] for $^{nat}\text{Pb}(p,x)^{196}\text{Au}$ and $^{nat}\text{Pb}(p,f)^{95}\text{Zr}$ (crosses). For $^{nat}\text{Pb}(n,x)^{196}\text{Au}$, the guess function used for the unfolding is given as broken line.

Cross sections cannot be directly calculated from these response integrals since the neutrons used are just "quasi mono-energetic" with only about 30 to 50% of the neutrons in the high-energy peak with a width of a few MeV. The neutron cross sections $\sigma(E)$ were extracted from production rates

P_i ($i = 1, \dots, n$) determined in a series of n irradiation experiments with different neutron energies by unfolding using the STAYS'L formalism [28, 29]. The unfolding needs a guess function to start with and which in our case was calculated using the ALICE-IPPE code [30]. But, the unfolding procedure is rather independent of the guess functions [31]. The advantage of this evaluation method is that it gives as results complete excitation functions together with their uncertainties and that evaluations can be repeated with improvements of the excitation functions as soon as new experimental information becomes available.

The exemplary results of this investigation given in Fig. 5 emphasize the importance to distinguish proton- and neutron-induced reactions leading to a residual nuclide. The excitation functions for the proton- and neutron-induced production mostly differ strongly with respect to shapes and absolute values. These figures also demonstrate the partially severe deviations of the final neutron excitation functions from the theoretical guess functions.

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